Plasma-Enhanced Combustion of Propane Using a Silent Discharge

L.A. Rosocha, Y. Kim, S. Stange, V. Ferreri (P-24), D.M. Coates (P-DO), D. Platts (P-22)

As a primary objective, researchers in P Division's Plasma Physics Group (P-24) want to minimize U.S. energy dependency on foreign resources through experiments incorporating a plasma-assisted combustion unit. Under this broad category, researchers seek to increase efficiency and reduce NO_x/SO_x and unburned hydrocarbon emissions in internal-combustion engines, gas-turbine engines, and burner units. To date, the existing lean-burn operations, consisting of a higher air-to-fuel ratio, have successfully operated in a regime where reduced NO_x/SO_x emissions are expected and have also shown increased combustion efficiency (less unburned hydrocarbon) for propane. By incorporating a lean-burn operation assisted by a non-thermal-plasma (NTP) reactor, the fracturing of hydrocarbons can result in increased power, combustion efficiency, and stability in the combustion system.

NTP units produce energetic electrons but avoid the high gas and ion temperatures involved in thermal plasmas. One NTP method, known as a silent discharge, allows free radicals to act in propagating combustion reactions, as well as intermediaries in hydrocarbon fracturing. Using NTP units, researchers have developed a fuel activation/conversion system that can decrease pollutants while increasing fuel efficiency, thus providing a path toward future U.S. energy independence.

Background

Combustion processes impact many aspects of modern life. They provide propulsion for automobiles, aircraft, and ships; generate electricity; and heat homes, water, and commercial buildings. Maximizing the efficiency of these combustion processes to conserve fuel and reduce pollution is of vital importance.

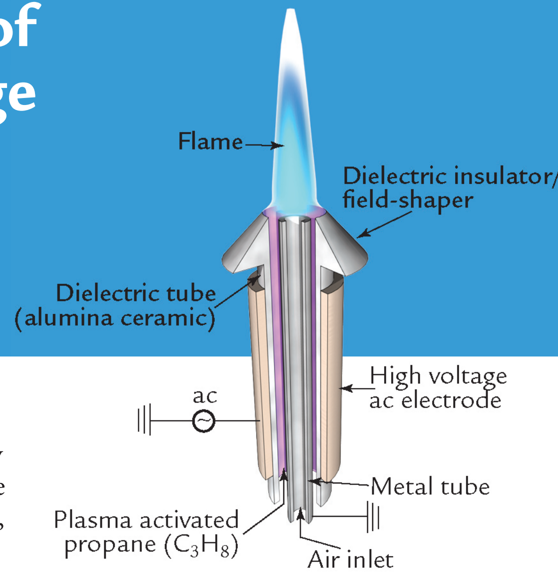

Figure 1. Schematic diagram of the experimental setup for blowout and combustion product studies, employing a coaxial DBD reactor.

Over the past five decades, many attempts have been made to improve combustion using an electric field, which can affect flame stability, flame propagation speed, and combustion chemistry.[1,2] However, the magnitude of the electric field in these experiments was insufficient to generate plasma. Thermal plasmas, which are usually less efficient and selective in directing electrical energy into the promotion of chemical reactions, have been applied to combustion over the past three or more decades with some success,[3] in particular, to convert air-fuel mixtures (into H_2 and CO),[4] to increase internal-combustion engine efficiency, and to reduce NO_x.

NTPs are potentially more useful tools for promoting combustion. In NTPs, the electrons are energetic ("hot"), whereas ions and neutral gases are near ambient temperature ("cool"), which results in little waste enthalpy (heat) being deposited in a process gas stream. Typical electron temperatures in such plasmas are at about a few electron volts, which is sufficient to break down the fuel and to produce free radicals.[5] We consider the silent electric discharge,[6] also known as a dielectric barrier discharge (DBD), as a very promising candidate for combustion enhancement. In 1983, Inomata *et al.*[7] demonstrated increases in flame speed when a DBD is applied upstream of a methane-air flame. More recent work performed by Cha *et al.*[8] showed that applying a DBD to the flame region results in a decrease in flame length and reduced soot formation.

Our new technology, based on NTPs, pre-treats fuels (not fuel-air mixtures) just prior to combustion.[9] In our technique, fuels are broken down (cracked) into smaller molecular fragments, boosted into reactive excited states, or made into "free-radicals." The "activated" fuel is then mixed with air and combusted. This technology allows for very "lean-burn" modes of combustion highly desirable for reduction of NO_x. "Proof of principle" has been demonstrated in experiments using propane as the fuel in a flame-based burner. We investigated the effects of the plasma on combustion by examining combustion stability under lean-burn conditions, observing increases in flame propagation speed by photograpy, and sampling and analyzing the gas residues from combustion.

Hypothesis for NTP Combustion Enhancement

Conventional propane-air combustion begins with spark ignition, whereby a spark thermally decomposes the propane-air mixture to produce free radicals and other reactive species. Burning then continues by the propagation of the reactive species generated by the heat of combustion. The overall combustion reaction rate is usually determined by how efficiently new reactive species are generated in the propagating flame front. However, the self-generation of reactive species is sometimes insufficient to sustain combustion under certain conditions, for example, during lean-burn operation.

NTP "activation" can be used to continuously convert atomized-liquid or gaseous fuels into reactive species, so that combustion does not rely on the self-generation of reactive species. The main possible mechanisms for fuel-cracking and fuel-activation (creation of more reactive species) are based on electron-impact processes, such as dissociation, dissociative ionization, vibrational excitation, and electronic excitation of the parent fuel molecule. Under an electron impact, propane is also ionized into multiple species, and these species then further fragment into smaller molecular ions.

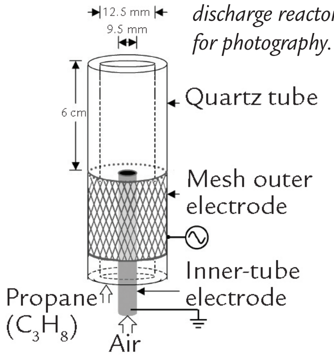

Figure 2. A coaxial silent discharge reactor, configured for photography.

Experimental Setup

We used two different NTP/DBD reactors for our investigations: one for lean-burn operation and exhaust-gas species determination and a second for flame-propagation observations. A schematic diagram of the first experimental setup[9] is shown in Figure 1. Air flows through a grounded tubular inner electrode (diameter of 0.96 cm). Propane (C_3H_8) flows through the annular gap between the inner electrode and an alumina ceramic tube (inner diameter of 1.9 cm). The ceramic tube is surrounded by a cylindrical metal outer electrode, which is powered by a high-voltage alternating-current (ac) transformer operated at about 450 Hz to match our propane DBD reactor. An NTP was formed in the propane stream, thus activating the fuel. The inner electrode is shorter than the ceramic tube, so there is a region (of variable length, but generally < 14 mm) where the fuel and air partially mix before being ignited. A ceramic nose-cone shapes the electric field at the end of the reactor to prevent arcing.

We used the reactor shown in Figure 2 for our flame-velocity observations.[10] In this experiment, the outer, high-voltage electrode was a piece of copper mesh. The mesh surrounded a quartz tube with an inner diameter of 12.5 mm. The inner electrode was a grounded stainless-steel tube having an outer diameter of 9.5 mm. Propane flowed in the annual region between the inner electrode and the quartz tube, while the air flowed down the center of the inner electrode. The ends of the electrodes and the end of the quartz tube were separated by a 6-cm mixing region. By using a relatively long mixing region, we were able to eliminate any effects of the electric field on the flame. The air- and propane-flow rates were set to 4.6 and 0.3 lpm, respectively, to fix the equivalence ratio at 1. At an equivalence ratio of 1, combustion is stoichiometric, or ideal, so the propane should be entirely consumed.

The power deposited into the plasma was measured using Lissajous diagram techniques (charge-voltage plot).[11] Other diagnostics included two thermocouples to measure inlet and outlet gas temperatures, a digital camera to take photographs of the flame, and a residual gas analyzer (RGA) equipped with a quadrupole mass spectrometer to measure the partial pressures of combustion by-products.

Experimental Results

Influence of Plasma on Flame Blowout Limits. We conducted blowout tests by holding the propane flow constant and increasing the air-flow rate until the flame blew out.[12] The blowout air-flow rate is an indicator of flame stability, and a high-blowout air-flow rate shows that combustion continues to occur under lean-burning conditions. Figure 3 shows the minimum blowout air-flow rates of an inverse, partially premixed flame for propane flow rates between 0.2 and 0.8 lpm. The number associated with each data point in the plot corresponds to the equivalence ratio ϕ,[13]

$$\phi = \frac{15.6 \times (\textit{propane flow rate})}{(\textit{air flow rate})}, \tag{1}$$

which is a standard measurement of combustion. In the absence of a plasma, the blowout limit of a propane flame increases with the propane flow rate and begins to saturate at a propane flow rate of 0.6 lpm. When 10-W discharge power is applied to the fuel, the blowout limit shows a large increase for low propane flow (and low equivalence ratio). However, the plasma benefit decreases as the propane flow increases, and for propane flow > 0.6 lpm, the blowout rate actually decreases in the presence of a plasma. This is not necessarily detrimental because low-equivalence-ratio systems show large decreases in pollutant production, especially NO_x, and are of great practical interest. In this experiment, the discharge power was held constant (10 W) while the propane-flow rate was increased. Thus, the discharge energy density ε,

$$\varepsilon = \frac{discharge\ power}{gas\ flow\ rate}, \quad (2)$$

deposited into the propane decreased as the propane flow rate increased. For example, at a propane flow rate of 0.3 lpm, the discharge energy density was equal to 2 kJ/l, whereas at a propane flow rate of 0.8 lpm, the discharge energy density fell to 0.75 kJ/l. Thus, the magnitude of the discharge energy density seems to affect the blowout limit of a propane flame. More experiments will be performed to correlate the combustion enhancement with the discharge energy density.

Combustion By-products. The concentrations of gaseous products of combustion were measured with the RGA. Mass fragments of particular interest are atomic masses 26 (C_2H_2), 27 (C_2H_3), 39 (C_3H_3), and 43 (C_3H_7). During operation, the flame was ignited and allowed to burn without plasma for two minutes. Then the power supply was turned on, and the activated fuel burned for two minutes. This procedure was repeated several times to test the repeatability of any enhanced combustion provided by the plasma.

Figure 4 shows typical data (with one mass fragment, M = 43 shown).[12] The partial pressure of the propane fragments decrease while water and carbon dioxide (both common hydrocarbon combustion products) increase when the plasma is turned on. The plateaus at the end of the traces are the result of extinguishing the flame—these are the partial pressures in the absence of any combustion. It is clear that the plasma significantly decreases the partial pressure of unburned hydrocarbons, indicating that propane is being burned more completely.

Flame Propagation Speed. The photographs displayed in Figure 5 were taken with a Canon PowerShot S45 digital camera with ISO 400, a focal length of 17.5 mm, an aperture of f/8.0, and a shutter speed of 0.8 s.[10] The images focus on the propane-air mixing region—the end of the quartz tube is visible at fiducial (1), and the purple glow at fiducial (2) is the edge of the outer electrode.

The progressively higher-power propane plasma's effect on the flame is shown in Figure 5. Figure 5(a) shows a propane-air flame in the absence of plasma. The application of even a relatively low-power 4-W plasma, as shown in Figure 5(b), improves the flame symmetry, a marker of stability. In both 5(a) and 5(b), the flame propagates upward only, although at an equivalence ratio of 1, it can theoretically propagate downward.[14] The latter indicates that the flame propagation rate is insufficient to overcome the upward flow of the propane-air mixture. When a 6-W propane plasma is created, as shown in Figure 5(c), the flame begins to propagate downward. As larger fields are applied to the propane gas, creating higher-power plasmas, downward propagation becomes increasingly pronounced, as shown in Figures 5(d)–(f). The changes in the flame's ability to propagate downward suggest that the flame-propagation rate increases with plasma power.

The flame propagates more quickly because it is igniting and burning faster. This combustion enhancement may result from the improved cracking of propane, the creation of reactive radicals, or

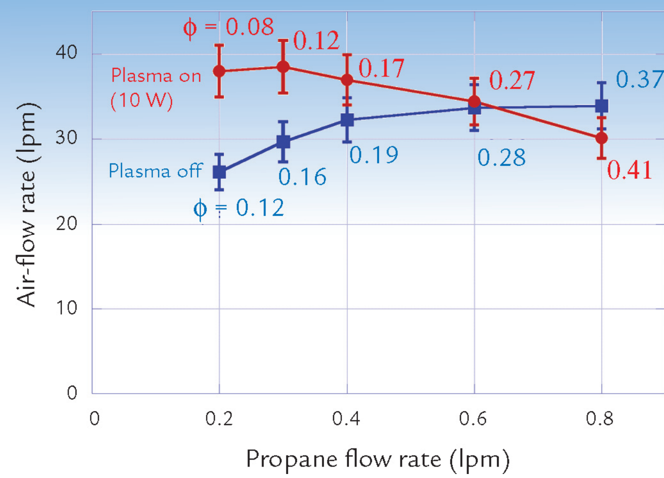

Figure 3. Effect of plasma on the blowout air-flow rate for propane flame. Combustion enhancement is indicated by the increment of the blowout limit when the plasma is on.

hydrogen generation. As discussed above, all of these factors likely play a role in combustion enhancement, but the relative importance of each is unknown. In the near future, we plan to use mixing regions of varying lengths to better understand the role of reactive radicals.

Conclusion

We have shown that silent-electrical-discharge-generated NTP can be used to activate propane fuel, significantly enhancing combustion in an activated propane-air mixture, as determined by mass spectrometric measurements of combustion-effluent gas concentrations. The plasma energy density required to achieve such enhancement is modest, of order 100's of J/std liter. Also, visual observations of activated propane-air flames indicate an increased spatial stability of the flame, increased blowout limits (leaner burn), and increased flame propagation speed. If applications to other fuels (e.g., gasoline, diesel, jet fuel) are successful, NTP-assisted combustion may prove to be highly beneficial to the energy needs of modern society.

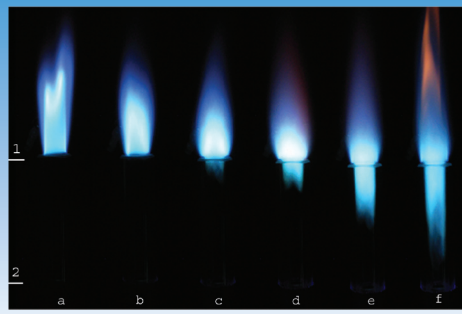

Figure 5. Increasing the plasma power causes an increase in flame propagation speed, indicated by the movement of the flame towards the electrodes. Figure (a) shows combustion without a plasma. The other images show the effect of plasmas having powers of (b) 4 W, (c) 6 W, (d) 8 W, (e) 10 W, and (f) 12 W. The numbers indicate regions of interest of the apparatus: (1) is the top of the quartz tube and (2) is the top of the outer electrode. The mixing region lies between them.

References

1. J. Lawton and F.J. Weinberg, *Electrical Aspects of Combustion* (Clarendon, Oxford, 1969).
2. H.C. Jaggers and A. von Engel, "The effect of electric fields on the burning velocity of various flames," *Combustion and Flame* **16**, 275–285 (1971).
3. F.J. Weinberg, Ed., *Advanced Combustion Methods* (Academic Press, London, 1986).
4. L. Bromberg, D.R. Cohn, A. Rabinovitch, and J. Heywood, "Emissions reductions using hydrogen from plasmatron fuel converters," *International Journal of Hydrogen Energy* **26**, 1115–1121 (2001).
5. L.A. Rosocha, "Processing of hazardous chemicals using silent electrical discharge plasmas," in *Environmental Aspects in Plasma Science*, W. Manheimer, L.E. Sugiyama, and T.H. Stix, Eds., (American Institute of Physics Press, Woodbury, New York, 1997).

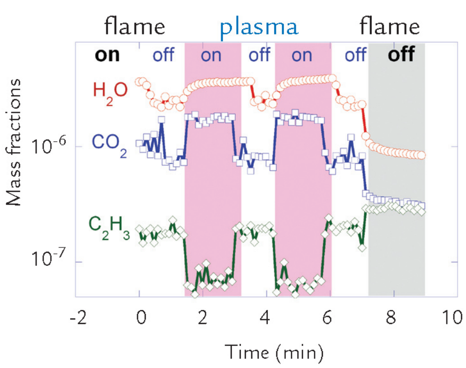

Figure 4. Mass spectrometer data for propane combustion fragments. Combustion enhancement is indicated by the reductions in unburned hydrocarbon and increases in water and carbon dioxide when the plasma is on.

6. U. Kogelschatz, "Dielectric-barrier discharges: Their history, discharge physics, and industrial applications," *Plasma Chemistry and Plasma Processing* **23**, 1–46 (2003).
7. T. Inomata, S. Okazaki, T. Moriwaki, and M. Suzuki, "The application of silent electric discharges to propagating flames," *Combustion and Flame* **50**, 361–363 (1983).
8. M.S. Cha, S.M. Lee, K.T. Kim, and S.H. Chung, "Interaction between a non-thermal plasma and a flame," in *Proceedings of the 30th International Conference Plasma Science*, Cheju, Korea, June 2–5, 2003.
9. L.A. Rosocha, D.M. Coates, D. Platts, and S. Stange, "Plasma-enhanced combustion of propane using a silent discharge," *Physics of Plasmas* **11**, 2950–2956 (2004).
10. S. Stange, Y. Kim, V. Ferreri, L.A. Rosocha, and D.M. Coates, "Flame images indicating combustion enhancement by dielectric barrier discharges," in *IEEE Transactions on Plasma Science* (to be published, February 2005).
11. T.C. Manley, "The electrical characteristics of the ozonator discharge," *Transactions of the Electrochemical Society* **84**, 83–95 (1943).
12. Y. Kim, S. Stange, and L.A. Rosocha, "Combustion enhancement of propane by dielectric barrier discharges," in *Proceedings of the 4th International Symposium on Non-Thermal Plasma Technology for Pollution Control and Sustainable Energy Development*, (Panama City Beach, Florida, May 10–14, 2004) pp. 221–225.
13. N.O. Nylund and A. Lawson, *Exhaust Emissions from Natural Gas Vehicles* (International Association for Natural Gas Vehicles, Sigma Group, Takapuna, New Zealand, 2000).
14. A.G. Gaydon and H.G. Wolfhard, *Flames: Their Structure, Radiation, and Temperature* (Chapman and Hall, London, 1970).

Acknowledgment

The authors wish to express their appreciation to Margaret Harris and Andrew Jensen (student interns) for technical assistance with this work and to P Division and TT Division for financial support for this project.